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Implementation of the Missing Aerosol Physics into LLNL IMPACT

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Implementation of the Missing Aerosol Physics into LLNL IMPACT

Principal Investigator: Catherine Chuang

I. INTRODUCTION

In recent assessments of climate forcing, the Intergovernmental Panel on Climate Change lists aerosol as one of the most important anthropogenic agents that influence climate. Atmospheric aerosols directly affect the radiative fluxes at the surface and top of the Earth's atmosphere by scattering and/or absorbing radiation. Further, aerosols indirectly change cloud microphysical properties (such as cloud drop effective radius) that also affect the radiative fluxes. However, the estimate of the magnitude of aerosol climatic effect varies widely, and aerosol/cloud interactions remain one of the most uncertain aspects of climate models today.

The Atmospheric Sciences Division has formulated a plan to enhance and expand our modeling expertise in aerosol/cloud/climate interactions. Under previous LDRD support, we successfully developed a computationally efficient version of IMPACT to simulate aerosol climatology. This new version contains a compact chemical mechanism for the prediction of sulfate and also predicts the distributions of organic carbon (OC), black carbon (BC), dust, and sea salt. Furthermore, we implemented a radiation package into IMPACT to calculate the radiative forcing and heating/cooling rates by aerosols. This accomplishment built the foundation of our currently funded projects under the NASA Global Modeling and Analysis Program as well as the DOE Atmospheric Radiation Program.

Despite the fact that our research is being recognized as an important effort to quantify the effects of anthropogenic aerosols on climate, the major shortcoming of our previous simulations on aerosol climatic effects is the over simplification of spatial and temporal variations of aerosol size distributions that are shaped by complicated nucleation, growth, transport and removal processes. Virtually all properties of atmospheric aerosols and clouds depend strongly on aerosol size distribution. Moreover, molecular processing on aerosol surfaces alters the hygroscopic characteristics and composition of aerosols. These processes, together with other physical properties (i.e., size, density, and refractive index), determine the atmospheric lifetime of aerosols and their radiative forcing. To better represent physical properties of aerosols, we adapted an aerosol microphysics model that simulates aerosol size distribution. Work toward this goal was done in collaboration with Professor Anthony Wexler of University of California at Davis. Professor Wexler's group has developed sectional models of atmospheric aerosol dynamics that include an arbitrary number of size sections and chemical compounds or compound classes. The model, AIM (Aerosol Inorganic Model), is designed to predict the mass distribution and composition of urban and regional particulate matter (*Sun and Wexler*, 1998a, b). This model is currently incorporated into EPA's Models-3 air quality modeling platform / CMAQ (Community Multiscale Air Quality) to test its performance with previous simulations of CMAQ over the continental US.

II. TCEHNICAL APPROACH

Over the six-month covered by this project, we modified AIM into a box model and successfully implemented it into the aerosol version of LLNL IMPACT model (T3A) (*Chuang et al*., 2002). This aerosol version contains a compact chemical mechanism for the prediction of sulfate, and also predicts the distributions of organic carbon (OC), black carbon (BC), dust, and sea salt. In an Eulerian flame of reference, the particle size and composition distribution is influenced by emission and deposition, condensation and evaporation, advection and diffusion, coagulation, nucleation, gravitational settling, and aerosol-phase chemical reactions. The general dynamic equation that describes the composition of an internally mixed aerosol over time is

$$\frac{\partial m(D_p,t)}{\partial t} = \left[\frac{\partial m(D_p,t)}{\partial t}\right]_{\text{emis-sion}} + \left[\frac{\partial m(D_p,t)}{\partial t}\right]_{\text{nucle-ation}} + \left[\frac{\partial m(D_p,t)}{\partial t}\right]_{\text{condensation/ evaporation}} + \left[\frac{\partial m(D_p,t)}{\partial t}\right]_{\text{coagu-lation}} + \left[\frac{\partial m(D_p,t)}{\partial t}\right]_{\text{advection/ gravitational settling}} + \left[\frac{\partial m(D_p,t)}{\partial t}\right]_{\text{chemical reaction}} + \left[\frac{\partial m(D_p,t)}{\partial t}\right]_{\text{diffusion}} \quad \text{Eq.(1)}$$

where $m(D_p, t)$ is the mass distribution such that $m(D_p, t)\delta D_p$ is the mass concentration of particles in the range $[D_p, D_p+\delta D_p]$. Ambient aerosol particles contain water and water-soluble inorganic compounds, elemental carbon, organic compounds and mineral dust. The sources of sea salt (NaCl), elemental carbon, dust and a portion of the organics are typically direct emissions. Most of the ammonium and nitrate, and the remaining organic compounds derive from gas-to-particle conversion processes. The particulate sulfate is derived from condensation of sulfuric acid (H_2SO_4) formed in gas phase reactions or from aqueous phase oxidation of SO_2 in clouds. The detailed representation for each process is given in *Wexler et al.* (1994).

Equation (1) was applied in conjunction with the treatments of physical process in IMPACT to predict both the composition and size distributions of atmospheric aerosols. The condensation and evaporation term is solved in AIM and the remaining processes (except for nucleation and coagulation) are solved using numerical methods available in IMPACT.

Nucleation and coagulation processes are currently not included in AIM, arguing that only a small fraction of aerosol mass is associated with homogeneous H_2SO_4-H_2O and H_2SO_4-HN_3-H_2O nucleation and coagulation is too slow to influence aerosol number distributions for particle diameters > 0.05 μm (*Zhang and Wexler*, 2002). While the growth of aerosols by condensation, in general, is being recognized as the leading process for modification of aerosol size distribution, homogeneous nucleation could be important in the upper troposphere where surface area of existing particles is relatively low (*Clarke*, 1993) or in regions recently scavenged by cloud (*Hoppel and Frick*, 1990) or in the proximity of clouds (*Hegg et al.*, 1990). We plan to revisit these issues to have complete physical processes as described by Eq.(1) in IMPACT if additional funding is available in the future.

III. RESULTS

We performed a full year simulation of IMPACT T3A/AIM plus a 3-month spin up from clean atmosphere using MACCM3 meteorology at resolution of 4º × 5º and 52 vertical layers. Model input includes emissions of primary aerosols of organic carbon (OC) and black carbon (BC) as well as aerosol precursors of SO_2, DMS, and H_2S from the Global Emissions Inventory Activity (GEIA) with both anthropogenic and natural origins. Emissions of sea salt and dust are calculated interactively in IMPACT as a function of local surface wind speed and other meteorological conditions (*Gong et al.*, 1997; *Ginoux et al.*, 2001).

Figure 1 presents the simulated annual aerosol mass distributions over east China, west Europe, and South America at σ levels of 1 (~ surface) and 0.7 (z ~ 3 km), respectively. Results are shown for simulations with 8 size sections. Significant regional variabilities in aerosol size distributions and compositions are noted, though these three regions are all characterized as high anthropogenic emissions of SO_2 and carbonaceous aerosols (OC + BC). In addition to anthropogenic aerosols dominant in fine particles, Asian dust and Mediterranean dust contribute a major portion of coarse particles over east China and west Europe, respectively. The size distribution of atmospheric aerosols not only depends on the initial size distribution of primary aerosols but also depends on the growth and removal processes. The growth mechanism is mainly associated with condensation and evaporation of condensable (e.g. H_2SO_4) and volatile (e.g. HNO_3 and H_2O) gas species with a rate strongly dependent on the composition at mass transfer, while the removal processes include gravitational settling, as well as dry and wet scavenging. These processes together with the short life time of aerosols result in large temporal and spatial variations in aerosol size distribution and compositions.

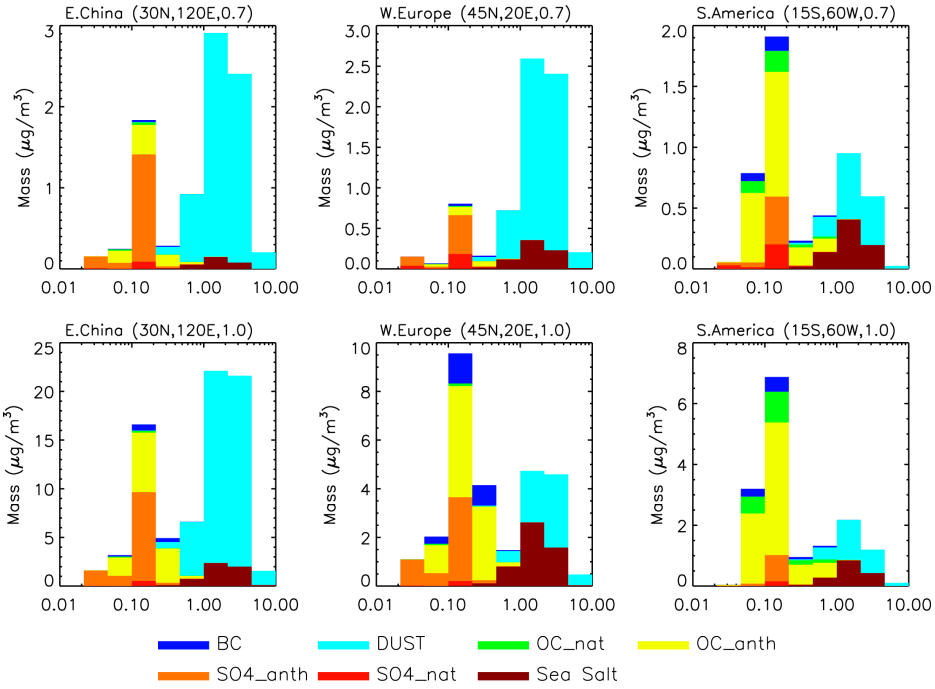

Figure 1. Simulated annual averages of aerosol mass distributions (μg/m^3) in different regions at σ = 1.0 and 0.7. Contribution from individual species is denoted by the color legend.

Figure 2 presents the corresponding aerosol number distributions. Concentrations of aerosols, in general, decrease by an order of magnitude between surface to 700 mb. Since the removal processes of aerosols are size-dependent, super-micrometer particles are removed from the atmosphere much more efficiently than sub-micrometer aerosols and narrow the width of aerosol size distribution.

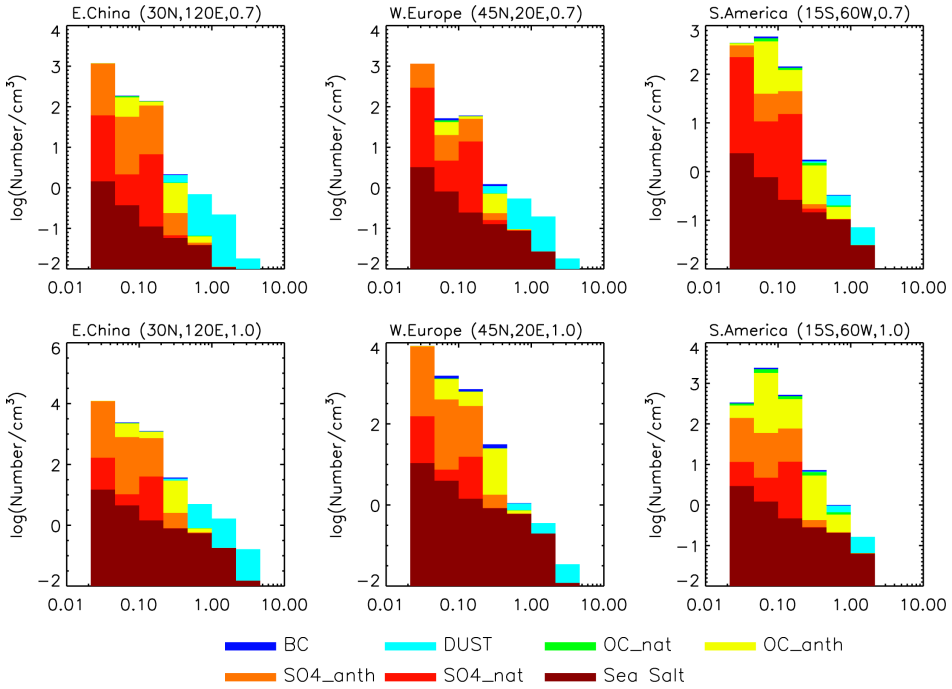

Figure 2. Same as Figure 1 but for number distributions ($1/cm^3$) assuming unit aerosol density.

We also examined the temporal variations of aerosol size distribution. Figure 3a shows the mass distributions over the west coast of California in January, May, and September at σ = 0.85 (z ~ 1.5 km), respectively. Seasonal variations are significant with low aerosol loading in January, high dust concentration from Asia in May, and high sulfate burden in September corresponding to stronger oxidation of SO_2 in summer time. To explore the sensitivity of aerosol representation to different number of size sections, Figure 3b presents results from 5 bins for comparison. We tested the cpu resource required for different number of aerosol size sections and found a 40% increase in cpu time for model with 8 aerosol bins than that with only 5 bins. Should the additional funding becomes available, we will validate the simulated aerosol size distributions and compositions with data available from in-situ campaigns as well as data retrieved from remote sensing instruments.

The size distribution of aerosols is critical to all climate influences. This work is aiming at reducing the forcing uncertainties associated with aerosol size distribution, chemical composition, and state of mixing, and would place the estimates of aerosol radiative forcings on a much more quantitative foundation than is available at present

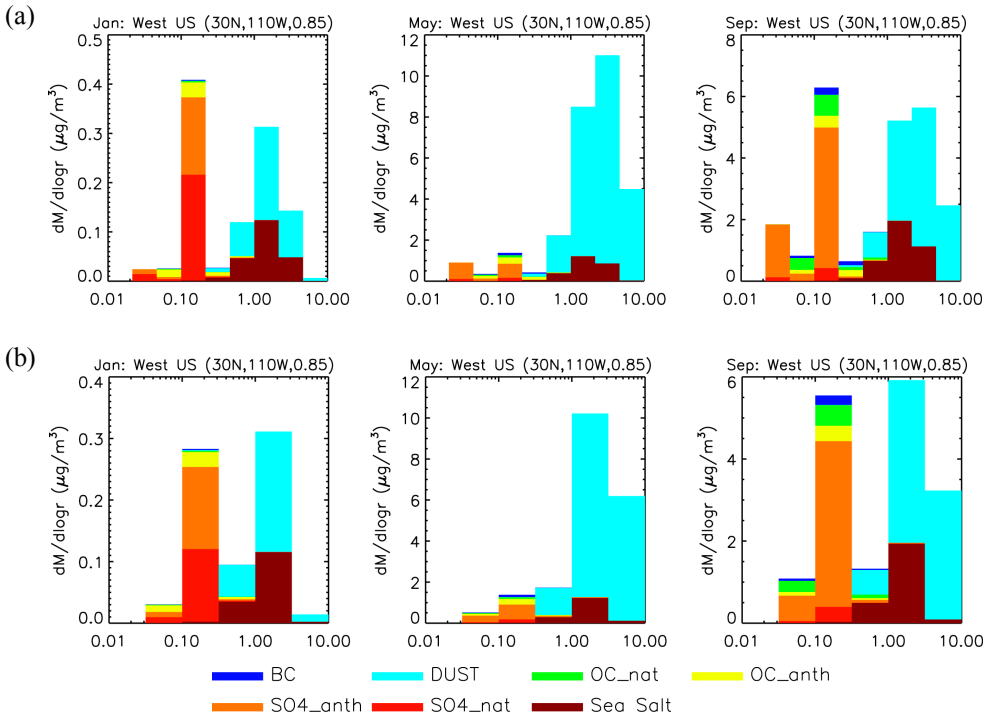

Figure 3. Simulated mass distributions over the west coast of California in January, May, and September at $\int = 0.85$ (z ~ 1.5 km) with (a) 8 size sections, and (b) 5 size sections.

IV. EXIT PLAN

The new model capability would place LLNL in a strong position to play major roles in two DOE programs. First is the DOE Atmospheric Radiation Measurement (ARM) Program, which aims to improve general circulation models and, in particular, the representation of cloud/radiation interactions in GCMs. Accurate treatments of aerosol physics in IMPACT are crucial to address the scientific issues of aerosol/cloud/radiation relationship on a much more quantitative foundation. Secondly, the DOE Atmospheric Science Program (ASP) with focus on aerosol radiative forcing. Linking the IMPACT model to an aerosol microphysics model will allow LLNL to play the central role in modeling aerosol radiative forcing. Another advantage in gaining expertise in this aerosol microphysics model is its applicability to models of all scales. This proposal focuses on its application to global issues of aerosol physics, but the model itself is equally suitable for regional and urban modeling efforts.

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VI. ACKNOWLEDGEMENTS

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